ARKIV FÖR FYSIK Band 15 nr 6

Communicated 12 November 1958 by MANNE SIEGBAHN and ERIK HULTHÉN

Light polonium isotopes from carbon ion bombardments of platinum

By HUGO ATTERLING and WILHELM FORSLING[1]

With 5 figures in the text

ABSTRACT

By bombardments of platinum with ^{12}C and ^{13}C ions in the Stockholm 225-cm cyclotron polonium isotopes of low mass numbers have been produced and their alpha-decay properties studied. One of the alpha activities found has been assigned to ^{203}Po with a particle energy of 5.48 MeV and a half-life of 45 ± 10 minutes. For ^{202}Po a half-life of 43 minutes has been obtained. This is considerably shorter than has been reported earlier.

Introduction

Somewhat different alpha-decay data and mass assignments have been reported for neutron-deficient polonium isotopes by different authors [1–8]. The data listed in Table 1 for ^{200}Po exemplifies this.

In order to examine the usefulness of heavy ion bombardments for studying neutron-deficient nuclides in this region a series of neon and carbon ion irradiations was carried out in the 225-cm cyclotron of the Nobel Institute of Physics. Thus, by use of neon ions and wolfram targets [6, 9] several low-mass polonium isotopes were found one of which was assigned to ^{196}Po. It was shown [9] to have a half-life of 1.8 minutes and to emit alpha particles of 6.13 MeV energy.

The present paper is concerned mainly with an investigation of polonium isotopes obtained by bombarding platinum with carbon ions. In addition, a study of the activities produced by carbon ions in iridium is briefly discussed, as this study is important for the question whether for instance possible impurities of iridium in the platinum can give rise to short-lived bismuth isotopes which may be confused with rapidly decaying polonium activities, when separated in fast procedures.

Experimental technique

The bombardments were made with internal beams and the ions utilized were $^{12}C^{4+}$ and $^{13}C^{4+}$. The gas used for the production of ^{13}C ions (CO_2) was enriched to about 60 per cent in the isotope ^{13}C.

---

[1] A report of this investigation was presented at the Conference on Reactions between Complex Nuclei, Gatlinburg, Tenn., U.S.A., May 5–7, 1958.

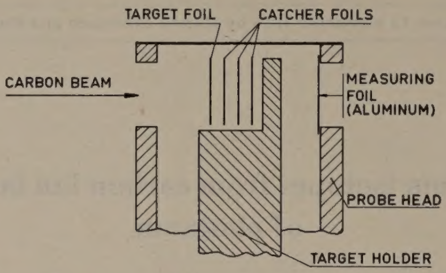

Fig. 1. Schematic drawing of target and catcher foil arrangement. The probe can be rotated 180° for bringing the measuring window into the beam.

For collecting the radioactive products the recoil technique was used. A schematic drawing of the target and catcher foil arrangement is shown in Fig. 1.

The platinum target was a circular foil, 8 mm in diameter and 10 μ thick (about 20 mg/cm^2). Catcher foils of Tygon (0.7–0.8 mg/cm^2) were placed close behind the target foil. All foils were attached to aluminium rings which were clamped to the target holder.

A similar arrangement was used for bombarding iridium. In this case the target consisted of an iridium film on a thin nickel backing. This target was prepared by painting an organic iridium solution onto a nickel foil. The solvent was evaporated and the foil ignited in an oven. The whole procedure was repeated many times in order to obtain a target layer of sufficient thickness. A circular disk of the foil was then mounted onto a supporting aluminium ring as mentioned above.

The probe could be withdrawn through an air lock and the target holder detached within about half a minute after the end of the bombardment.

The bombardment of platinum with ^{12}C ions was performed at a radius of 90 cm and the one with ^{13}C ions at 85 cm. The bombarding times were about half an hour each. The nominal energy corresponding to the radii used was approximately 130 MeV in both cases. No determination of the beam spectrum was made in connection with the present investigation, but an estimate of the energy distribution could be made on the basis of foil experiments performed under similar cyclotron conditions. According to this estimate the majority of the high-energy ions (^{12}C ions as well as ^{13}C ions) had energies in the range 90–120 MeV with the distribution maximum somewhere in the region 100–120 MeV. When bombarding platinum the intensities of the beams reaching the target holder with energies above 90 MeV were approximately 3×10^{-2} μA for ^{12}C ions and 2×10^{-2} μA for ^{13}C ions. A detailed description of heavy ion acceleration in the Stockholm cyclotron and the bombarding technique used, will be reported elsewhere [10].

In several polonium preparations different methods for the chemical separations were used [11]. As the platinum target gave no activity from ^{203}At or ^{205}At after carbon bombardments there were no interfering impurities of gold present. From the iridium experiments it was found that the alphaactivity due to bismuth isotopes was very low. Experiments showed that bismuth was not evaporated under the radiofrequency heating conditions used for the volatilization of polonium isotopes from one platinum foil to another. Based on these results the following rapid method for preparing the polonium sample was used.

Immediately after the bombardment the catcher foil of Tygon was dissolved by a few drops of acetone on a platinum plate and the solution dried under an infrared lamp. The plate was then placed in an evaporation device on the top of which a water-cooled platinum counting disk served as catcher for the polonium, evaporated from the first-mentioned plate. This procedure was the only one used which was rapid enough for the study of the most short-lived activities mentioned above.

For the counting of the alphaparticles and the determination of their energies an ionization chamber combined with a 50-channel pulse analyzer [12] was used. Measurements could be started within about 4 minutes after the end of the bombardment.

Results and discussion

As is well known, most nuclides in the region studied in the present investigation decay predominantly by electron capture. This causes some difficulties in the alpha particle energy determinations especially in the measurements of the activities induced by carbon ions in iridium targets. The results of these measurements will be published elsewhere. However, in connection with the polonium studies it was, as mentioned above, of value to know if short-lived bismuth activities which could interfere with the polonium measurements were likely to be formed. The energies and half-lives given by Neuman and Perlman [13] for the neutron-deficient bismuth isotopes were confirmed.

The present authors hope to be able to make definite mass assignments of those isotopes, which are assumed to be ^{199}Po and ^{200}Po, by isotope separation. So far, the short half-lives and the relatively small alpha intensities have made such work unfeasible.

An alpha-particle energy spectrum obtained from a ^{13}C bombardment is shown in Fig. 2. It should be pointed out that the peak energies have been determined from several spectra, as generally the number of experimental points in a single spectrum was too low, due to the channel widths used, for an accurate energy determination. The alpha-particle energies and the half-lives found for polonium isotopes of mass numbers from 196 to 204 in three different studies are summarized in Table 1. The isotopes in the range from ^{197}Po to ^{204}Po have been produced and

Table 1. Alpha-particle energies and half-lives of neutron-deficient polonium isotopes.

Po mass number	Karraker, Ghiorso and Templeton, 1951 [4]		Rosenblum and Tyrén, 1954 [5]		This work and ref. 9[1], 1956–1958	
	E_α (MeV)	$t_{\frac{1}{2}}$	E_α (MeV)	$t_{\frac{1}{2}}$	E_α (MeV)	$t_{\frac{1}{2}}$
196					6.13	1.8 min
197			6.040	~4 min	6.03	4 min
198			5.935	~6 min	5.93	~7 min
199			5.846	~11 min	5.86	12 min
200	5.84	11 min	5.770	~8 min	5.77	10 min
201	5.70	18 min	5.671	~17 min	5.68	18 min
202	5.59	52 min	5.575	~55 min	5.58	43 min
203	—	47 min	—	—	5.48	45 ± 10 min
204	5.37	3.8 hr	5.370	~3.8 hr	5.38	3–4 hr

[1] The data for ^{196}Po are from ref. 9.

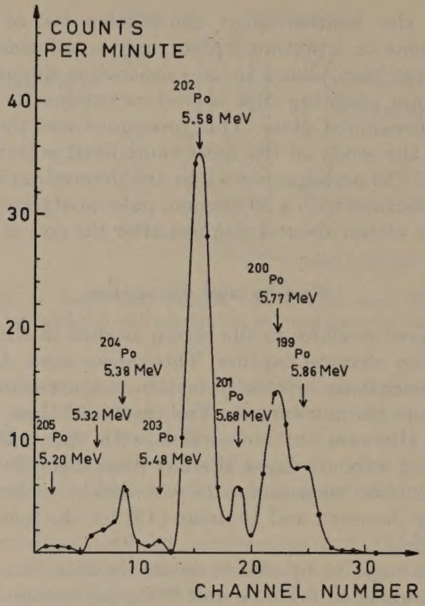

Fig. 2. Alpha-particle energy spectrum of neutron-deficient polonium isotopes taken 4–11 minutes after the end of the ^{13}C-bombardment. The weak activity at about 5.20 MeV has been ascribed to ^{205}Po for which earlier investigations [4] have given an alpha-particle energy of 5.21 MeV.

investigated in the present experiments. In addition to the activities attributed to these isotopes, a weak alpha-particle group which is assumed to be due to ^{205}Po has been observed.

The errors in the energy data given in Table 1 by the present authors are estimated to be ± 0.03 MeV and the errors in the half-lives in general to be about ± 10 per cent. An exception from this is the error in the half-life of ^{203}Po which is estimated to be ± 10 minutes because of the low intensities obtained.

In the alpha-particle energy spectrum the ^{203}Po peak is very small. However, it was easily reproduced and a weak indication of this peak is also possible to observe in the energy spectra given by the two other above-mentioned research teams [4, 5]. As will be shown below, this activity could not be due to the growth of ^{199}Bi from ^{199}Po.

The half-lives found by the present authors for the different neutron-deficient polonium isotopes generally agree very well with those earlier found. However, an exception has to be made for ^{202}Po. In this case the half-lives found by us is about 10 minutes shorter than the values given earlier. The measurements were repeated with the same result. The decay curve is shown in Fig. 3.

The activity assumed to be due to ^{205}Po was obtained in a poor yield under the experimental conditions used. At about 5.32 MeV the energy spectrum given in Fig. 2 seems to indicate an activity which we have not been able to identify.

In Table 2 the yields of the different polonium isotopes from the ^{12}C and ^{13}C bombardments are given as relative saturation alpha activities. Owing to the choice

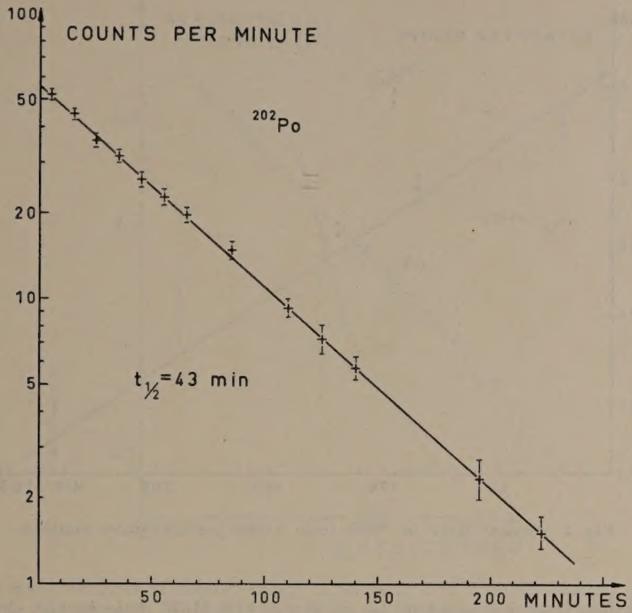

Fig. 3. Decay curve of ^{202}Po from alpha particle pulse analysis.

of bombarding radii these bombardments could be expected to give approximately the same excitation energies. The polonium isotopes are, of course, formed by nuclear reactions with all the different stable platinum isotopes constituting natural platinum. A rough estimate of the excitation of the compound nucleus formed by the capture of a ^{12}C or ^{13}C ion by any of these isotopes has been made in the following way. The

Table 2. Alpha saturation activities of polonium isotopes produced by ^{12}C and ^{13}C bombardments of platinum in percentage of the ^{202}Po activity obtained in each bombardment.

Po mass number	Relative activities	
	^{12}C bombardment	^{13}C bombardment
197	17	0
198	10	0
199	22	15
200	95	40
201	30	9
202	100	100
203	5	4
204	25	50
205	5	3

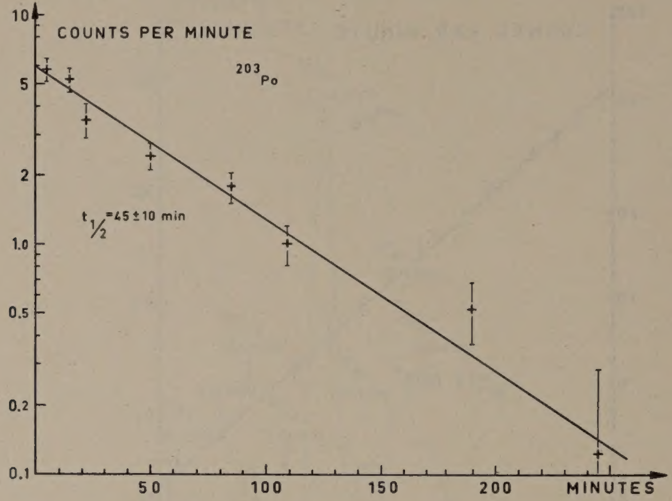

Fig. 4. Decay curve of ^{203}Po from alpha particle pulse analysis.

energy peak has been assumed to be at about 110 MeV. Due to the short range in platinum of the polonium atoms formed, only the polonium activity produced in a thin surface layer on the rear side of the platinum target has been collected by the catcher [14]. Therefore, the ions causing the reactions considered in this paper have been slowed down by about 20 mg/cm^2 platinum which corresponds to a loss in energy of approximately 15 MeV. Taking this into account the excitation energies of the compound nuclei of interest is estimated to be in the range 70–75 MeV (for ions reaching the target foil with energies of 110 MeV). In this calculation the atomic masses tabulated by Martin [15] have been used. Information on the most probable neutron evaporation reactions at different excitation energies can be obtained from the calculations of (p,xn) cross sections by Jackson [16]. According to Jackson's curves for the realtive probabilities for neutron evaporation from heavy compound nuclei, as those of polonium, the most probable number of neutrons emitted at the excitation energies estimated above would be 6 or 7. We obtain consistent results by estimating the nuclear temperature on the basis of the formula and data given by Blatt and Weisskopf [17] and using neutron binding energies from Martin's tables. It should be noted, that the carbon ion energy distribution may have had a tail in the range 120–130 MeV. In the target layer of interest in our discussion ions with incident energies of, for instance, 125 MeV will cause excitations in the range 85–90 MeV. From Jackson's results we find that a $(C,8n)$ reaction would be the predominating one in this case.

The isotope ^{198}Bi which is expected to be formed by electron capture from ^{198}Po obtained in the ^{12}C-bombardment has a half-life of about 7 minutes and an alpha-particle energy of 5.83 MeV [13]. This activity will contribute to the alpha peak of ^{199}Po. The genetic relationships involved are the following [18]:

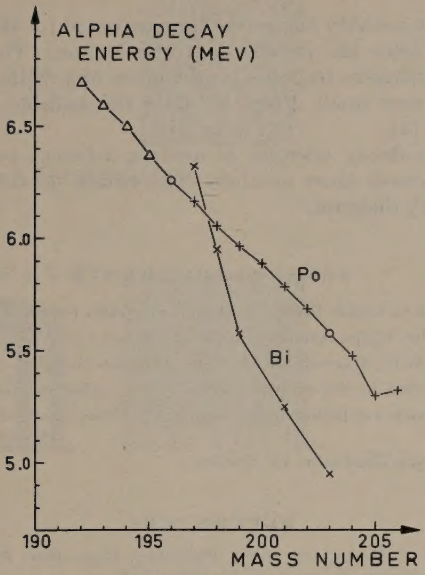

Fig. 5. Alpha-decay energies of neutron-deficient polonium and bismuth isotopes plotted versus mass numbers. +, polonium isotopes with earlier reported alpha decay properties [1–5]. ○, polonium isotope energy values from the present work and ref. 9 (see Table 1). △, polonium isotope energy values given by P. A. Tove [7]. ×, bismuth isotopes.

$$\begin{tikzcd} {}^{198}\mathrm{Po} \arrow[r, "\mathrm{EC}", "t_{\frac{1}{2}} = 7\ \mathrm{min.}"'] \arrow[d, "\alpha"'] & {}^{198}\mathrm{Bi} \arrow[r, "\mathrm{EC}", "t_{\frac{1}{2}} = 7\ \mathrm{min.}"'] \arrow[d, "\alpha"', "5 \times 10^{-2}\,\%"] & {}^{198}\mathrm{Pb} \arrow[r, "\mathrm{EC}", "t_{\frac{1}{2}} = 25\ \mathrm{min.?}"'] & {} \\ {}^{194}\mathrm{Pb} & {}^{194}\mathrm{Tl} & & \end{tikzcd}$$

Even if the EC/α ratio for ^{198}Po is of the same order as that of ^{198}Bi (2×10^3), the growth of ^{198}Bi should hardly have been observed in the decay curves of ^{199}Po or ^{200}Po from our experiments. Certainly the ratio is much smaller.

According to well-known laws of radioactive decay the maximum activity of ^{198}Bi will in this case appear 10 minutes after the purification of the polonium. However, no growth was observed.

A similar calculation was made for ^{199}Bi formed from ^{199}Po according to the following scheme:

$$\begin{tikzcd} {}^{199}\mathrm{Po} \arrow[r, "\mathrm{EC}", "t_{\frac{1}{2}} = 12\ \mathrm{min.}"'] \arrow[d, "\alpha"'] & {}^{199}\mathrm{Bi} \arrow[r, "\mathrm{EC}", "t_{\frac{1}{2}} = 25\ \mathrm{min.}"'] \arrow[d, "\alpha"', "10^{-2}\,\%"] & {}^{199}\mathrm{Pb} \arrow[r, "\mathrm{EC}", "t_{\frac{1}{2}} = 90\ \mathrm{min.}"'] & {} \\ {}^{195}\mathrm{Pb} & {}^{195}\mathrm{Tl} & & \end{tikzcd}$$

As the alpha-particle energy of ^{199}Bi is 5.47 MeV it should appear together with ^{203}Po in the energy spectrum provided the value of EC/α ratio for ^{199}Po is not too low. In fact, the decay curve for ^{203}Po in Fig. 4 gives no significant indication of such a growth of ^{199}Bi alpha activity before the time expected for the maximum intensity of ^{199}Bi (24 minutes after the purification of polonium). The half-life of 45 ± 10 minutes found also indicates that the contribution of a ^{199}Bi alpha activity to the ^{203}Po peak must be very small. From EC data the half-life of ^{203}Po is known to be about 47 minutes [4].

In Fig. 5 the alpha-decay energies of neutron-deficient polonium and bismuth isotopes are plotted versus mass numbers. The circles show the positions of ^{203}Po and ^{196}Po in the energy diagram.[1]

ACKNOWLEDGEMENTS

The authors would like to thank Professor Manne Siegbahn for his kind interest in this work. Thanks are also due to Dr. Björn Åström for the electronic equipment put at our disposal and to Dr. John Milsted, A.E.R.E., Harwell, for the loan of the ion chamber used in these experiments. Further thanks are extended to the cyclotron crew and to other members of the technical staff for their assistance. The work was facilitated by supply of a ^{13}C-compound from A.E.R.E., Harwell.

Nobel Institute of Physics, Stockholm 50, Sweden.

REFERENCES

1. D. H. Templeton, J. J. Howland, and I. Perlman, Phys. Rev. *72*, 758 (1947).
2. E. L. Kelly and E. Segrè, Phys. Rev. *75*, 999 (1949).
3. D. G. Karraker and D. H. Templeton, Phys. Rev. *81*, 510 (1951).
4. D. G. Karraker, A. Ghiorso, and D. H. Templeton, Phys. Rev. *83*, 390 (1951).
5. S. Rosenblum and H. Tyrén, Compt. rend. *239*, 1205 (1954).
6. H. Atterling, W. Forsling, and B. Åström, Proceedings of the International Conference on Nuclear Reactions, Amsterdam 1956; Physica, *XXII*, 1193 (1956).
7. P. A. Tove, Arkiv för fysik *13*, 549 (1958).
8. R. G. Harris, Private communication.
9. H. Atterling, W. Forsling, and B. Åström, To be published in Arkiv för fysik.
10. H. Atterling, To be published in Arkiv för fysik.
11. G. Oelsner and W. Forsling, Arkiv för kemi *11*, 349 (1957).
12. B. Åström, Arkiv för fysik *12*, 215 (1957).
13. H. M. Neumann and I. Perlman, Phys. Rev. *78*, 191 (1950).
14. Compare, for instance, the study of astatine ranges by R. B. Leachman and H. Atterling, Arkiv för fysik *13*, 101 (1957).
15. C. N. Martin, Tables Numériques de Physique Nucléaire. Gauthier-Villars, Paris 1954.
16. J. D. Jackson, Can. J. Phys. *34*, 767 (1956).
17. J. B. Blatt and V. F. Weisskopf, Theoretical Nuclear Physics. John Wiley & Son, New York 1952, p. 372.
18. D. Strominger, J. M. Hollander, and G. T. Seaborg, Table of Isotopes, Rev. Mod. Phys. *30*, 585 (1958).
19. H. Atterling, W. Forsling, and B. Åström, ORNL-2606, Proceedings of the Conference on Reactions Between Complex Nuclei, Gatlinburg, Tenn., U.S.A., May 5–7, 1958, p. 248.

---

[1] Unfortunately, due to a mistake, the decay energy values of Po$^{192-195}$, which were kindly communicated to us by Dr. P. A. Tove, were marked in the same way as our own values in Fig. 4 of our report in the Proceedings of the Conference on Reactions Between Complex Nuclei, Gatlinburg, Tenn., U.S.A., May 5–7, 1958 [19].

Tryckt den 25 februari 1959

Uppsala 1959. Almqvist & Wiksells Boktryckeri AB